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54 Propanil dispersible granule formulation.

57 A dispersible granular formulation has been developed which contains at least 60% propanil herbicide. This formulation also provides excellent suspensibility and dispersibility characteristics and resists attrition.

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This invention relates to novel dispersible granule formulations of the herbicide propanil and processes for their preparation.

5 Water dispersible granular pesticide formulations are known. These formulations are desirable because they avoid the use of potentially toxic solvents and permit the use of easily-disposable paper containers or water soluble containers. Potential exposure of pesticide applicators and the general public to the pesticide or solvent is thereby reduced.

10 Typical dispersible granular pesticide formulations are described for example in GB-A-1,433,882, EP-A-0,252,896 and US-A- 3,920,442. GB 1,433,882 describes a process for preparing dispersible granules by blending premilled, water-insoluble, active ingredient, dispersing agents, disintegrating agent and wetting agents in an aqueous suspension. The aqueous mix is extruded to form granules which are then dried to yield the final product. US-A-3,920,442 describes water dispersible pesticide aggregates containing 5 to 95% by weight of pesticide. The aggregates are prepared by contacting the finely divided solid ingredients in a fluidized bed with a fine spray of water or a solution of the binder-dispersant followed by drying.

15 In attempts at making pesticide granules, it has often been found that granules or agglomerates prepared from the formulated wettable powders of the art using well-known agglomerating techniques and using water as the agglomerating agent, are not easily dispersible in water. On the other hand, agglomerates which are readily water-dispersible are often not sufficiently resistant to attrition and form a fine dust fraction upon handling and shipping. If conventional binders are added to make the granules strong, then they are not dispersible in water. Techniques such as tabletting, extrusion and rolling which involve high-pressure compaction of moistened mixtures containing finely divided pesticides, diluents, binder and dispersant, as described in US-A-20 3,617,246, lead to dense pellets, tablets, plates, and rods which are subsequently dried and crushed. These techniques have also been used to form granules containing up to 50 % of active pesticide, but the resulting granules are not rapidly or completely water dispersible and are not suitable for use in preparing sprayable suspensions.

25 Propanil herbicide is N-(3,4-dichlorophenyl)propionamide. Dispersible granular (DG) formulations are designated propanil DG or dry flowable (DF). The percentage active ingredient in the formulation is sometimes indicated, as for example, propanil 80 DG or propanil 60 DG.

30 A DG herbicide composition designed for dispersion in a liquid carrier should ideally have a high content of active material, should be readily dispersible in the carrier and should then form a dispersion which is as stable as possible, requiring the minimum of subsequent agitation for homogeneity. The liquid carrier will, of course, for convenience normally be water.

35 Low-melting solids such as propanil present an especially difficult problem in the preparation of a dispersible granule formulation. The low-melting solid, herein defined as melting below 100° C., tends to melt or become sticky during or subsequent to the grinding process which is a necessary step in preparing dispersible granules. EP-A-0,252,896 describes a possible solution to this problem which requires micro-encapsulation of low-melting pesticides prior to granulation. However, microencapsulation involves additional processing steps and adds to the cost of the overall formulation.

40 In the case of the herbicide propanil, it has not heretofore been possible to produce a dispersible granular product which combines the features of a high active ingredient content and good suspensibility and dispersant properties, as well as resistance to attrition-thereby avoiding the formation of a dust.

45 Commercial formulations of propanil dispersible granules are available, but these products have deficiencies in that the active ingredient content is relatively low and/or they fail to provide adequate suspension or dispersibility characteristics or are not resistant to attrition.

We have now discovered a process for producing dispersible granule formulations of propanil herbicide which can provide, granules containing more than 60% active ingredient which also have good suspensibility and dispersibility characteristics, and which resist attrition.

50 Accordingly in one aspect this invention provides a process for producing a dispersible propanil granule comprising:

- 55 a) combining one or more surfactants with propanil and milling to a particle size of less than 20 microns to form a pre-mix;
- b) adding less than 25 percent by weight water and optionally a wetting agent to said pre-mix and mixing until a paste is obtained;
- c) granulating said paste thereby producing granules; and
- d) drying said granules.

55 A further aspect of the invention provides a dispersible granule comprising at least 60 percent by weight propanil, said granule having a suspensibility of at least 70 percent, preferably at least 80%, and being dispersible in fewer than 15 cylinder inversions.

By granular form is meant granules substantially all of which have a mean particle size of at least 1 mm.

i.e a particle size much larger than the mean particle size of a powder, the mean particle size of which is measured in microns.

Preferably, the granule has less than 0.3% by weight of particles less than 45 microns in diameter after 10 minutes of attrition.

5 A more preferred granule of this invention is a dispersible granule comprising at least 80% propanil, said granule having a suspensibility of at least 65%, preferably at least 80% and dispersibility in less than 15 cylinder inversions.

10 In addition to the active ingredient, propanil, the mixture to be formed into dispersible granules will generally contain one or more surfactants and, optionally, flow enhancing agents, dispersants, wetting agents and defoaming agents.

15 The suspensibility test was performed according to the procedure of the Collaborative International Pesticides Analytical Council (CIPAC) Handbook, Vol. 1, Ed. G. R. RAW (1970), Method Number MT 15.1. Standard hard water (342 ppm as calcium carbonate) was prepared according to CIPAC method MT 18.1.4., also known as Army Hard Water. Suspensibility is measured as a 0.9% to 2.0% weight/volume dispersion in standard hard water.

20 Dispersibility is measured by placing one gram of the dispersible granules in 100 ml of 342 ppm hardness water and repeatedly slowly inverting the test cylinder until the material is completely dispersed. Preferably, dispersion should be complete in 15 or fewer cylinder inversions.

25 "Attrition" as used in this application is defined as the reduction in particle size which occurs when propanil granules are shaken with steel balls as described in Example 1(c)(3).

The term "surfactant" is used in the broad sense to include materials which may be referred to as emulsifying agents, dispersing agents and wetting agents, and the surfactant component may comprise one or more surfactants selected from the anionic, cationic and nonionic type.

30 Examples of surfactants of the anionic type include soaps, salts of aliphatic monoesters of sulfuric acid such as sodium lauryl sulfate, salts of sulfonated aromatic compounds, for example sodium dodecylbenzene sulfonate, sodium, calcium or ammonium lignosulfonate or butylnaphthalene sulfonate, and a mixture of the sodium salts of diisopropyl- and triiso-propynaphthalene sulfonates. Suitable agents of the non-ionic type include, for example, the condensation products of ethylene oxide with fatty alcohols such as oleyl alcohol or cetyl alcohol, or with alkyl phenols such as octyl phenol, nonyl phenol and octyl cresol. Other non-ionic agents are the partial esters with ethylene oxide and the lecithins and phosphorylated surfactants such as phosphorylated ethylene oxide/propylene oxide block copolymer and ethoxylated and phosphorylated styryl substituted phenol.

35 Preferably the surfactant component will comprise at least one wetting agent such as those selected from alkyl naphthalene sulfonates, alkylaryl polyoxyethylene ammonium sulfonates phosphate esters, sulfosuccinates and nonionics such as tridecyl alcohol ethoxylate; and/or at least one dispersing agent such as those selected from the group of naphthalene sulfonates, lignosulfonates, polyacrylates and phosphate esters.

Typically the total surfactant component will comprise from 0.1 to 25% and preferably from 1 to 15% by weight of the dry weight of the composition.

40 In the context of this specification a dispersing agent is a surfactant which facilitates the dispersion of the pesticide particles when the product is added to a liquid, for example water. The dispersing agents used are preferably water-soluble ones. Examples of dispersants preferred for the dispersible granule formulations of this invention include: Tamol 731®, Polyfon® H, Polyfon O, Reax® 88B, Morwet® D-425, Reax 45DA, Polyfon T, Polyfon F, Polyfon H, Lignosol™ XD-65, Reax 45L, Reax 85A, Reax 910, Polyfon OD, PC-825 Polyfon T. Most preferred dispersants are: Reax 85A and Polyfon H.

45 Examples of surfactants preferred as wetting agents for the dispersible granule formulations of this invention include Morwet® B, Morwet EFW, Sellogen® DFL, Morwet IP, Igepon® AC-78, Igepon T-77, Aerosol OT-8, and Surfactant XN-45S. Most preferred wetting agents are Morwet B and Surfactant XN-45S.

50 All surfactants act as dispersing agents to some degree, and also to some degree as wetting agents; most surface-active agents are, however, more efficient in one capacity than the other. The worker of ordinary skill in the formulation art can select a surfactant most suitable for the purpose in view.

55 Small particles of a low-melting solid such as propanil often tend to stick together thereby causing flow problems in processing the material. Flow aids such as clays or silica particles may be used to minimize these problems. Flow aids preferred for the propanil dispersible granular include HiSil® 233, Wessalon® 50S, Cab-O-Sil® M-5, Wessalon S, Berden® Clay, and Microcel® E. Most preferred are HiSil 233 and Wessalon 50S. The flow aid content of the dispersible granule may vary from 0 to 10% and preferably from 1 to 8%.

60 Addition of a silicon-containing antifoaming agent is desirable in the processing and use of propanil dispersible granules. Defoaming agents may be used in amounts of 0.1% to 5%; a preferred range is about 0.2% to 1.0%. The preferred defoaming agent is Mazu DF-1300.

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8 Disintegrants, which are water soluble, organic compounds such as starch or sugar or inorganic salts such as sodium acetate or sodium bicarbonate, are sometimes used in dispersible granule formulations. See GB 1,433,882 for example. We have found that these disintegrants have adverse effects on the propanil dispersible granules of this invention. Formulations containing disintegrants were found to be more dusty and disperse less effectively than the granules of this invention.

9 "Dust" is herein defined as particles having a diameter less than 45 microns. A solid having less than 0.3% by weight dust is defined as "non-dusty" while a solid with more than 4% dust is defined as "very dusty". Solids with 0.3-4% dust are defined as "dusty".

10 Dust content and resistance to attrition of the propanil dispersible granules of this invention are minimized by forming the granules by the preferred mode of extrusion.

11 The most preferred composition of this invention comprises by weight percent: propanil 80%; dispersant, Reax 85A, 9.2%; flow aid, HiSil, 4.0%; wetting agent, surfactant XN-45S, 2.0%; defoaming agent, Mazu DF-1300, 0.5%; and water, less than 1%.

12 Examples of dispersant, wetting agents, flow aids and defoaming agents useful in this invention are shown in Table I.

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TABLE I

| | <u>Dispersants</u> | <u>Producer</u> | <u>Chemical Type</u> |
|----|-----------------------|--|--|
| 5 | Tamol® 731 | Rohm and Haas Co. Philadelphia, PA 19105 | Sodium carboxylate polyelectrolyte |
| 10 | Polyfon® H | Westvaco Chemicals | Aliphatic and aromatic sulfonated lignin |
| | Polyfon F | | |
| | Polyfon T | | |
| | Polyfon O | | |
| | Polyfon OD | | |
| 15 | Reax® 88B | | |
| | Reax 45DA | | |
| | Reax 45L | | |
| | Reax 85A | | |
| | Reax 910 | | |
| 20 | Lignosol XD-65 | Reed Lignin, Inc. 81 Holly Hill Lane Greenwich, CT 06830 | Sodium lignosulfonate |
| 25 | <u>Wetting Agents</u> | | |
| | Morwet® B | DeSoto, Inc. 2001 N. Grove Fort Worth, TX 76113 | Sodium n-butyl naphthalene sulfonate |
| 30 | Morwet EFW | | Naphthelene sulfonate |
| | Morwet IP | | Sodium diisopropyl naphthalene sulfonate |
| 35 | Sellogen® DFL | Diamond Shamrock 350 Mt.Kemble Ave. Morristown, NJ 07960 | Alkyl naphthalene sulfonate |
| 40 | <u>Wetting Agents</u> | <u>Producer</u> | <u>Chemical Type</u> |
| | Igepon®AC-78 | GAF Corp. 140 W. 51st St. New York, NY 10020 | Sodium cocyl isethionate |
| 45 | Igepon T-77 | | Sodium methyl aleoyl taurate |
| | Aerosol OT-B | American Cyanamid | Sodium dioctyl sulfo succinate |
| 50 | Surfactant XN-45S | Rohm and Haas Co. Philadelphia, PA 19105 | Ammonium alkyl/aryl polyoxyethylene sulfate |

Flow Aids

| | | | |
|----|----------------|--|----------|
| 5 | HiSil 233 | PPG Industries One Gateway Center Pittsburgh, PA 15222 | Silica |
| 10 | Wessalon® 505 | Degussa Corp. Rt.46, Hollister Rd. Teterboro, NJ 07608 | Silica |
| | Wessalon S | | Silica |
| 15 | Cab-O-Sil® M-5 | Cabot Corp. Boston, MA 02110 | Silica |
| 20 | Barden® Clay | J.M. Huber Corp. Rt.#4 Macon, GA 30201 | Clay |
| 25 | Microcel® E | Johns-Mansville P.O. Box 5108 Denver, CO 80217 | Silicate |

Defoaming Agents

| | | | |
|----|--------------|--|---------------------|
| 30 | Mazu DF 1300 | Mazer Chemicals 3938 Poreti Drive Gurnee, IL 60031 | Silicone and Silica |
|----|--------------|--|---------------------|

35 The dispersible granules of this invention are generally prepared by milling one or more surfactants, combined with an amount of propanil sufficient to achieve at least 60% active ingredient in said granules, to a particle size of less than 20 microns, preferably less than 15 microns and more preferably less than 10 microns, thereby forming a pre-mix; adding to said pre-mix less than 25% water, based on the total weight of the composition optionally adding a wetting agent and mixing until a paste is obtained; granulating said paste; and drying the granules thus produced.

40 The term "pre-mix" as used herein refers to a mixture of active ingredient, plus optionally dispersant, flow aid and defoaming agent; the above ingredients are generally milled to a partial size of 3 to 15 microns, preferably about 8 to 9 microns to form the pre-mix.

45 The final composition is prepared from the pre-mix by adding water, optionally containing a dissolved wetting agent, thus forming a "paste", then blending, agglomerating and drying.

Preferred compositions of pre-mix and final composition are shown below.

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| <u>Function</u> | <u>Raw Material</u> | <u>Premix %</u> | <u>Final Composition %</u> |
|-------------------|-----------------------|-----------------|----------------------------|
| Active ingredient | Propanil | 85.9 | 83.3 |
| Dispersant | Reax 85A | 9.5 | 9.2 |
| Flow aid | HiSil 233 | 4.1 | 4.0 |
| Wetting agent | Surfactant XN-455 | ----- | 2.0 |
| Defoaming agent | Mazu DF-1300 Water | 0.5 ----- | 0.5 1.0 |

Agglomeration or granulation may be accomplished by any operable means such as tabletting, pan agglomeration, or extrusion. Extrusion is the preferred method.

Suspensibility of the granule is proportional to the amount of water added to the premix prior to extrusion granulation. In the case of however, addition of too much water will cause sticking of the extrudate as it exits the extruder. The relationship of water content of premix and suspensibility is shown in the table below.

| <u>Parts Water per 100 parts premix</u> | <u>% Suspensibility</u> |
|---|-------------------------|
| 8 | 63.1 |
| 14 | 78.4 |
| 18 | 84.6 |

The preferred amount of water to be added to the premix is 18 to 20 parts per 100 parts of premix.

After the granulation/agglomeration step, the dispersible granules are dried. For storage stability, it is important to reduce the residual water to at least 2% and preferably below 1%. High drying temperatures are injurious to the product. The preferred drying temperature is less than 60° C. and more preferably less than 40° C. Drying may be accomplished by any suitable drying means which supplies inert gas at a controlled temperature. A two stage fluid bed dryer is preferred.

A preferred process for producing the propanil dispersible granules of this invention comprises:

- (a) milling a mixture of propanil, dispersant and flow aid to a particle size between 3 and 15 microns;
- (b) adding a wetting agent dissolved in 12-20% water (based on the total weight of ingredients) to the milled mixture of step (a) and mixing until a homogeneous, extrudable paste is obtained;
- (c) extruding the paste obtained in step (b);
- (d) drying the extruded granules at a temperature of less than 60° C. to a moisture content of less than

The following examples describe the preparation of the propanil dispersable granules of this invention and

— 10 —

Preparation of Saponin-80-PC

a) Preparation of Procanil RA DG Premix

Technical propanil (97%) was melted by heating at 110°C. for 24 hours. The molten material was then poured into aluminum foil lined trays to a depth of about one inch. After cooling at room temperature for 24 hours, the solid was broken up and milled in a coffee mill. The ground material classified as follows:

| <u>Mesh Size</u> | <u>Weight %</u> |
|--------------------|-----------------|
| Larger than 4 mesh | 14.9 |
| 10 to 4 mesh | 40.5 |
| 10 to 20 mesh | 29.3 |
| 20 to 50 mesh | 8.3 |

5 The coarse milled propanil was blended with other formulation ingredients in a Marion mixer (Mfg. for Rapid
10 Machinery Co., Marion Mixer, Iowa by Texas Div. Tranter Inc. Old Burk Road, Wichita Falls, TX USA) in the
following ratio of ingredients:

| | <u>Weight %</u> |
|-----------------|-----------------|
| 15 Propanil | 85.9 |
| Reax 85A | 9.5 |
| HiSil 233 | 4.1 |
| 20 Mazu DF-1300 | 0.5 |
| Total | 100 |

25 and mixed for 20 minutes.

25 The above blend was transferred to a Bantam micropulverizer, (obtainable from Mikropul, 10 Chantaur Road, Summit, NJ 07901, USA, fitted with a 0.42 inch (1.1 cm) screen and automatic feed; the grinding chamber was cooled with dry ice. The mean particle size of micropulverized product was 43.8 microns.

30 The micropulverized product was then air milled in an 8" horizontal (pancake) jet mill, Fluid Energy Processing and Equipment Co., 153 Penn Energy, Hatfield, PA 19440. An Accu-Rate feeder, (Accurate Feeder, 746 E. Milwaukee Street, White Water, WI 53190, USA) was fitted with a 2" screw feed nozzle positioned to deliver micropulverized material to the air mill. High pressure nitrogen was used for both feeding and grinding to maintain an oxygen concentration below 10%. The ground material was collected in the air bag.

Air Mill Operating Conditions

| | |
|-------------------------|---------|
| Nitrogen feed pressure | 65 psig |
| Nitrogen grind pressure | 60 psig |
| Accurate-rate setting | 100 |

40 Under these conditions the milling rate was 8-10 lbs. (3.6-4.5 kg)/hour. The mean particle size was 9.1 microns. This product constituted propanil 80 DG premix.

b) Preparation of propanil 80 DG

45 Propanil premix (98.0 parts by weight), Surfactant XN-45S (80% aqueous solution, 3.33 parts by weight) and water (16.67 parts by weight) were mixed in a Kitchen Aid mixer, (Hobart, Kitchen Aid Div. Troy, Ohio, USA) for approximately 2-3 minutes.

50 The mixture was transferred to a KAR 130 extruder, (Tsu Tsui Rikagaku Kitai Co., Ltd., Japan) fitted with a 1.0 mm screen. After extrusion, the extrudate was air dried to a moisture content of 1-2% at a temperature of less than 40° C.

C.) Measurement of Physical Properties.

1. Suspensibility (variation of CIPAC method in duplicate)

One gram of propanil DG was placed in a glass-stoppered graduated cylinder containing 99 ml of Army Hard Water at 25°. The cylinder was inverted 30 times over a period of 90 seconds and allowed to stand for

30 minutes. The bottom 10 ml was separated, evaporated and dried for 48 hours at 60°C; and the resulting residue was used to calculate suspensibility by the following formula.

$$\% \text{ suspensibility} = [1 - \text{weight of residue}] \times 111$$

5 2. Dispersibility (in triplicate)

One gram propanil DG was added to 100 ml of Army Hard Water in a 100 ml glass-stoppered graduated cylinder. The cylinder was inverted until the DG was completely dispersed and the number of inversions was recorded.

10

10 3. Product Attrition Measurement

Ten 3/8 inch (1cm) steel balls and 50 g propanil DG were placed in the bottom pan of 8 inch (20 cm) diameter stainless steel sieves. The pan was shaken on a Ro-top sieve shaker with a hammer tapper for 10 minutes.

13

The steel balls were removed from the pan and the granules were transferred to the top of the nested sieves in the order of 20, 60, 100, 200, 325 mesh and bottom pan. The nested screens were inserted into the Ro-top and shaken for 15 minutes. The amount of residue on each sieve and bottom pan was determined. The weight percent of each fraction was determined by sieving before and after the attrition test.

20

20 4. Residual Moisture loss (in duplicate)

Approximately 5 grams DG was weighed to the nearest 0.01 g in a preweighed pan, and dried 24 hours (± 1 hour) at 60°C. at approximately 15 mm Hg pressure.

25

The percent moisture loss was measured by weighing the residue.

30

30 5. Disintegration/Suspensibility (specified in GB 1,433,882 on pg. 4)

One gram DG was added to 100ml Army Hard Water in a centrifuge tube and inverted 15 times slowly. The tube was then placed in a 30°C. water bath and readings (ml of sediment) were taken at 2, 5, 30 minutes.

After 30 minutes, suspension poured through a 120 mesh screen. The screen washed with 1 liter water and residue weight noted.

Results of the above measurements were as follows:

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| | | |
|----|----------------------|--------|
| 5 | % Suspensibility | 88.4 |
| | | 88.8 |
| | (average) | (88.6) |
| 10 | Dispersibility | 13 |
| | | 14 |
| | | 14 |
| | (average) | (14) |
| 15 | % Moisture loss | 1.1 |
| | | 1.5 |
| | (average) | (1.3) |
| 20 | Disintegration/Susp. | |
| | 2 min (<trace*) | 0.05ml |
| | 5 min (<0.1 ml*) | 0.1ml |
| 25 | 30 min (<0.3 ml*) | 0.3ml |
| | Screen residue | 0 |

* suggested target.

30 Results of Attrition Test

An increase of 2.4% particles below 840um (20 mesh) occurred following the attrition test. This increase was confined to particles within the range of 250um to 74um. No increase of percent particles below 44um occurred. Initial percentage below 44um was 0.2%. Data for the study are given in the following table.

| 35 | Part. Cut | Sieve | Attrit. | |
|----|----------------------|------------------|-----------------|--------------------|
| | | Test (Before) | Test (After) | Differ. |
| 40 | Through a 325 mesh | <44um | 0.20 | 0 |
| | Retained on 325 mesh | 44/74 | 0.16 | -0.02 |
| | Retained on 200 mesh | 74/149 | 0.08 | 0.10 |
| 45 | Retained on 100 mesh | 149/250 | 0.02 | 1.19 |
| | Retained on 60 mesh | 250/840 | 0.10 | 1.13 |
| | Retained on 20 mesh | >840um | <u>99.44</u> | <u>97.04</u> -2.40 |
| | | | 100.00% | 100.00% |

50 EXAMPLE 2

A number of formulations of propanil DG were prepared with active ingredient content ranging from 60.4% to 90.2%. The procedure for preparing these granular formulations followed the general procedure of Example 1, except that Monwet B, Polyfon H and Barden Clay were substituted for Surfactant XN-45S, Reax 85A and HISII 233, respectively. No antifoam agent was used. The results are shown in the Table II below.

TABLE II

| <u>Ingredients as WT. %</u> | | 60.4 | 65.4 | 70.4 | 75.4 | 75.4 | 80.2 | 85.2 | 90.2 |
|-----------------------------|---------------------------|------|------|------|------|------|------|------|------|
| 5 | Propanil Tech | 60.4 | 65.4 | 70.4 | 75.4 | 75.4 | 80.2 | 85.2 | 90.2 |
| 10 | Morwet B | 2.0 | 2.0 | 2.0 | 2.0 | 2.0 | 2.0 | 2.0 | 2.0 |
| 15 | Polyfon H | 10.0 | 10.0 | 10.0 | 10.0 | 21.1 | 16.2 | 11.1 | 6.0 |
| 20 | Barden Clay | 26.5 | 21.3 | 16.2 | 11.1 | 0 | 0 | 0 | 0 |
| | Total | 100 | 100 | 100 | 100 | 100 | 100 | 100 | 100 |
| | <u>Suspensibility (%)</u> | 91.7 | 100 | 100 | 100 | 100 | 92.7 | 64.5 | 14.0 |

EXAMPLE 3Comparison with Competitive products

1. Comparison of propanil DG with formulations incorporating disintegrants as described in British Patent 1,433,882.

20 Propanil 80DG was prepared with 0, 2 and 4 percent of the disintegrants sodium bicarbonate and sodium acetate. Various physical properties were measured using air dried, extruded product. Use of disintegrants, at either level, worsened dispersibility in water although there was some slight improvement in suspensibility with 2 percent sodium bicarbonate or 4 percent sodium acetate after 2 and 5 minutes. Samples containing sodium acetate required additional water in the wetting step to allow proper extrusion.

Procedure:

40 Using Propanil 80DG premix (milled but not extruded), a control and alternative extruded formulations containing 2 and 4 percent disintegrants were made. These were prepared according to the procedure of Example 1 and all compositions used 2 percent (solids) Surfactant XN-45S in the wetting step at the rate of 18:100 water to premix. The complete compositions were:

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| | <u>Composition</u> | <u>Propanil DG</u> | <u>GB 1.433.882 Disintegrants</u> | | | |
|----|--|--------------------|-----------------------------------|----------|----------|----------|
| | | | <u>Control</u> | <u>1</u> | <u>2</u> | <u>3</u> |
| 5 | Propanil Premix | 98.0 | | 96.0 | 94.0 | 96.0 |
| 10 | Surfactant XN-45S (60%) | 3.33 | | 3.33 | 3.33 | 3.33 |
| 15 | Water | 16.67 | | 16.67 | 16.67 | 16.67 |
| 20 | Sodium Bicarbonate ¹ | - | | 2.0 | 4.0 | - |
| 25 | Sodium Acetate ¹ | - | | - | - | 2.0 |
| 30 | | 118.00 | | 118.00 | 118.00 | 118.00 |
| 35 | Additional water added to facilitate extrusion | 0 | | 2.5 | 3.4 | 4.0 |
| 40 | | | | 6.0 | | |

The composition of propanil premix in the above table was:

| | | <u>Weight %</u> |
|----|--------------|-----------------|
| 25 | Propanil | 85.9 |
| 30 | HiSil 233 | 4.1 |
| 35 | Mazu DF-1300 | 0.51 |
| 40 | Reax 85 A | 9.5 |
| 45 | Total | 100.0 |

The premix and disintegrants, sodium bicarbonate or sodium acetate were mixed together in a Kitchen Aid mixer for approximately 1 minute. The surfactant/water solution was added while mixing and

Anhydrous powder

allowed to knead for 2-3 min. Initial compositions with disintegrants which were formulated with the same concentration of water as the control were too dry for extrusion (extrusion process did not form noodles). Additional water was added as indicated in the above table and kneading continued for an additional 1-2 min. The sample was transferred to the small KAR 130 extruder and extruded through the 1.0mm screen. The extrudate was air dried overnight and various measurements were taken.

Measurements:

Suspensibility, dispersibility, residual moisture, and disintegration/suspensibility (as specified in GB 1.433.882, pg. 4) were determined as described in Example 1. Results are shown in Table III.

Table III

Propanil DG GB 1,433,882 Disintegrates

| Control | 1 | 2 | 3 | 4 |
|---------|---|---|---|---|
|---------|---|---|---|---|

| | | | | |
|-----------------------|--------|--------|--------|--------|
| % Sodium bicarbonate | 0 | 2 | 4 | 0 |
| % Sodium acetate | 0 | 0 | 0 | 2 |
| g additional water | 0 | 0 | 0 | 4 |
| % Suspensibility | 88.4 | 90.2 | 89.7 | 88.0 |
| | 88.8 | 89.6 | 90.0 | 87.7 |
| (average) | (88.6) | (89.9) | (89.8) | (87.8) |
| Dispersibility (ave.) | 13 | 16 | 17 | 21 |
| | 14 | 16 | 17 | 21 |
| | 14 | 16 | 18 | 23 |
| (average) | (14) | (16) | (17) | (22) |
| % Moisture loss | 1.1 | 1.9 | 2.0 | 2.2 |
| | 1.5 | 1.9 | 1.9 | 2.2 |
| (average) | (1.3) | (1.9) | (1.9) | (2.2) |
| Disintegration/Susp. | | | | |
| 2 min (<trace*) | 0.05ml | trace | 0.15ml | 0.05ml |
| 5 min (<0.1ml*) | 0.1ml | trace | 0.15ml | 0.07ml |
| 30 min (<0.3ml*) | 0.3ml | 0.25ml | 0.3ml | 0.3ml |
| Screen residue | 0 | 0 | 0 | 0 |

*suggested target

The products of GB 1,433,882 containing disintegrants were rated dusty while the control granules of this invention were rated non-dusty.

2. Comparison of Propanil 80 DG with commercial products.

Cedar Chemical Corp., 5100 Poplar, Memphis, TN 38137 produces a 50% propanil dispersible granule. Terra International, Inc., Terra Centre 600 Fourth Street, Sioux City, IA 51101, produces a 60% propanil dispersible granule.

Propanil 80 DG was compared with commercial products from Cedar and Terra. Suspensibility was measured initially and after storage at 40° C and 54°C for 1 to 4 weeks. The results are shown in Table IV.

The Cedar product was rated very dusty having approximately 5% by weight of particles less than 45 microns in diameter after attrition. The Terra product was rated dusty. In contrast, two lots of the granules of this invention had 0.184% and 0.239% by weight of particles less than 45 microns in diameter after attrition and were rated non-dusty.

TABLE IV

| Mfg. | % Propanil | Initial | % Suspensibility | | | | 4 Weeks | |
|-------------------|------------|---------|------------------|------|---------|------|-----------|------|
| | | | 1 week | | 2 weeks | | 40°C 54°C | |
| | | | 40°C | 54°C | 40°C | 54°C | | |
| Cedar | 50 | 29 | 35.4 | 16.0 | 20.7 | 14.6 | 27.5 | 13.8 |
| Terra | 60 | 41.0 | 21.4 | 16.1 | 27.4 | 7.0 | 14.5 | 6.6 |
| Propanil DG 80 #1 | 89.0 | 91.0 | 88.5 | 88.9 | 88.8 | 89.0 | 88.3 | |
| Propanil DG 80 #2 | 89.6 | 91.2 | 89.2 | 90.0 | 89.3 | 87.4 | 88.1 | |
| Propanil DG 80 #3 | 91.2 | 91.3 | 88.7 | 90.8 | 87.8 | 90.6 | 86.5 | |
| Propanil DG 80 #4 | 91.7 | 90.3 | 86.0 | 87.9 | 83.3 | 88.2 | 86.0 | |

25 Samples #1 - #4 of this invention were prepared by the procedure of Example 1.

Sample #4 used Morwet B instead of Surfactant XN-45S as the wetting agent.

30 Although the invention has been described in regard to its preferred embodiments, which constitute the best mode presently known to the inventors, it should be understood that various changes and modifications as would be obvious to one having ordinary skill in this art may be made without departing from the scope of the invention which is defined in the claims.

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Claims

1. Process for producing a dispersible propanil granule comprising:
 - combining one or more surfactants with propanil and milling to a particle size of less than 20 microns to form a pre-mix;
 - adding less than 25 percent by weight water and optionally a wetting agent to said pre-mix and mixing until a paste is obtained;
 - granulating said paste thereby producing granules; and
 - drying said granules.
2. Process according to claim 1 wherein the amount of propanil employed is such that the final granules formed have a propanil content of at least 60% by weight, preferably at least 80%.
3. Process according to claim 1 or 2 wherein granulating step (c) is preferred by extruding the paste.
4. Process according to any preceding claim wherein one or more of a dispersant, a flowing and a defoaming agent are added during step (b).
5. Process according to any preceding claim comprising the steps of:
 - milling a mixture of propanil, dispersant and flow aid to a particle size between 3 and 15 microns;
 - adding a wetting agent dissolved in 12-20 percent by weight water to the milled mixture of step (a) and mixing until a homogeneous, extrudable paste is obtained;

- c) extruding said paste to form granules; and
- d) drying said extruded granules at a temperature of less than 60°C, preferably less than 40°C to a moisture content of less than 2%, preferably less than 1%.

5 6. A dispersible granule comprising at least 60 percent by weight propanil, and having a suspensibility of at least 70 percent, preferably at least 80%, and being dispersible in fewer than 15 cylinder inversions.

7. A dispersible granule according to claim 6 comprising at least 65 percent, preferably at least 80% by weight of propanil.

10 8. Dispersible granule according to claim 6 or 7 having less than 0.30 weight percent of particles which are less than 45 microns in diameter after 10 minutes of attrition as hereinbefore defined.

15 9. A dispersible granule according to any one of claims 6 to 8 further comprising a dispersant, preferably a sulfonated lignin, and/or a flow aid, preferably a silica compound, and/or a wetting agent, preferably a naphthalene sulfonate or polyoxyethylene sulfate, and/or a defoaming agent, preferably a silicone or silica compound.

20 10. A dispersible granule according to any one of claims 6 to 8 which is substantially free of disintegrant.

11. A dispersible granule according to any one of claims 6 to 10, made by a process according to any one of claims 1 to 5.

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| DOCUMENTS CONSIDERED TO BE RELEVANT | | | CLASSIFICATION OF THE APPLICATION (Int. Cl. 5) |
|--|--|----------------------------------|---|
| Category | Citation of documents with indication, where appropriate, of relevant passages | Relevant to claim | |
| X | MO-A-8 900 079 (ICI AUSTRALIA OPERATIONS PROPRIETARY) ■ page 1, line 2 - line 5 * ■ page 2, line 16 - page 3, line 10 * ■ page 3, line 22 - line 24 * ■ page 4, line 12 - line 13 * ■ page 5, line 34 - page 6, line 7 * ■ page 7, line 28 - page 8, line 12 * ■ page 8, line 25 - line 28 * ■ page 10, line 24 - page 11, line 11 * ■ examples * --- GB-A-1 399 005 (CIBA-GEIGY) ■ page 1, line 8 - line 11 * ■ page 2, line 14 - line 21 * ■ page 2, line 56 - line 85 * ■ page 2, line 98 - line 106 * ■ page 3, line 18 - line 52 * ■ examples * --- FR-A-2 215 171 (IMPERIAL CHEMICAL INDUSTRIES) ■ page 1, line 3 - line 7 * ■ page 1, line 37 - page 2, line 26 * ■ page 3, line 29 - line 39 * ■ page 4, line 11 - line 16 * ■ page 5, line 1 - line 11 * ■ page 5, line 34 - page 6, line 3 * ■ claims 1,8,9 * GB-A-1 433 682 --- EP-A-0 135 963 (NORDEUTSCHE AFFINERIE) ■ page 1, paragraph 1 * ■ page 3, paragraph 1 * ■ page 5, paragraph 2 -paragraph 4 * ■ claims 1,6 * --- | 1-11 | A01N37/22 //(A01N37/22, 25:14) |
| | The present search report has been drawn up for all claims | 1-9,11 | TECHNICAL FIELDS SEARCHED (Int. Cl. 5) A01N |
| | | 1-11 | |
| Name of search | | Date of completion of the search | Examiner |
| THE HAGUE | | 06 FEBRUARY 1992 | W. Lenars |
| CATEGORY OF CITED DOCUMENTS | | | |
| X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : co-written disclosure P : intermediate document | | | |
| I : theory or principle underlying the invention E : earlier patent document, not published on, or after the filing date D : document cited in the application L : document cited for other reasons R : member of the same patent family, corresponding document | | | |



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| DOCUMENTS CONSIDERED TO BE RELEVANT | | | CLASSIFICATION OF THE APPLICATION (Int. CL.5) | | | | | | |
|--|--|---|---|-----------------|----------------------------------|------------------|-----------|------------------|-----------|
| Category | Citation of document with indication, where appropriate, of relevant passages | Relevant to claims | | | | | | | |
| A | <p>US-A-3 657 446 (K. A. E. BLACKMORE)</p> <ul style="list-style-type: none"> * column 1, line 23 - line 26 * * column 2, line 33 - line 42 * * column 3, line 64 - column 4, line 7 * * example 2 * <p>---</p> <p>FR-A-2 595 544 (KAO)</p> <ul style="list-style-type: none"> * page 1, line 1 - line 8 * * page 1, line 35 - page 2, line 16 * * page 2, line 33 - page 3, line 23 * * page 4, line 20 * * page 5, line 13 - page 6, line 22 * * page 12, line 21 - page 13, line 9 * <p>---</p> | 1-11 | | | | | | | |
| | | | TECHNICAL FIELDS SEARCHED (Int. CL.5) | | | | | | |
| | | | | | | | | | |
| <p>The present search report has been drawn up for all claims</p> <table border="1" style="width: 100%; border-collapse: collapse;"> <tr> <td style="width: 33%;">Place of search</td> <td style="width: 33%;">Date of completion of the search</td> <td style="width: 34%;">Name of examiner</td> </tr> <tr> <td>THE HAGUE</td> <td>06 FEBRUARY 1992</td> <td>W. Lomers</td> </tr> </table> | | | | Place of search | Date of completion of the search | Name of examiner | THE HAGUE | 06 FEBRUARY 1992 | W. Lomers |
| Place of search | Date of completion of the search | Name of examiner | | | | | | | |
| THE HAGUE | 06 FEBRUARY 1992 | W. Lomers | | | | | | | |
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